Supporting Information for:

## Dynamic Assembly of DNA with Polylysine Mediated by Electric Energy

Lin Niu, Xuyan Yang, Xiaocui Zhu, Yudan Yin, Wei Qu, Jihan Zhou, Meiping Zhao\*, Dehai Liang\*

Beijing National Laboratory for Molecular Sciences and the Key Laboratory of Polymer Chemistry & Physics of Ministry of Education, College of Chemistry & Molecular Engineering, Peking University, Beijing 100871, P. R. China

## Method

**Chip electrophtores:** Salmon testes DNA (~2000 base pairs) and FITC-PLL (M<sub>W</sub>: 15000-30000 Da, FITC percentage 0.3%-1%) were purchased from Sigma-Aldrich (St. Louis, MI, USA) and used as received. Ultrapower<sup>TM</sup> DNA dye is purchased from Bioteke (Beijing, China). Milli-Q water (18.2 M $\Omega$ ) is used in all the experiments. Polyvinylpyrrolidone(PVP) is purchased from Sinopharm Chemical Reagent Co. Ltd (Beijing, China). The runner buffer is 0.2 × phosphate buffer. The concentrations of DNA and PLL are 5.0×10<sup>-5</sup> g/mL and 1.0×10<sup>-4</sup> g/mL, respectively, in 0.2×phosphate buffer. The mobility of free DNA is 2.2×10<sup>-4</sup> cm<sup>2</sup>/(V.s), similar to the value reported in literature. <sup>1</sup> The mobilities of PLL is about 2.8 × 10<sup>-4</sup> cm<sup>2</sup>/V\*s. <sup>2</sup>

Chip electrophoresis is conducted on a ZDMCI6-1 Microfluidic Chip Detection System (Hang-zhou Syltech Technology Co., Ltd.) equipped with an inverted microscope (Olympus, IX71). Designed double-cross-channel glass microchips were purchased from Dalian Institution of Chemical Physics, Chinese Academic of Science, China. The length of the separation channel is 17 mm long and all the other channels are 6 mm to the separation channel. All channels are 25  $\mu$ m in depth and 80  $\mu$ m in width. Reservoirs of 3 mm in diameter are drilled into the cover plate at the end of each channel. The chip is pretreated sequentially by 98% H<sub>2</sub>SO<sub>4</sub> for 10 minutes, H<sub>2</sub>O for 10 minutes, 1 M NaOH for 2 hours, and H<sub>2</sub>O for 10 minutes. 1% PVP in 0.2× phosphate buffer treat the channel for 10 minutes before use to suppress the electro-osmotic flow and to decrease the absorption of PLL.

Atomic Force Microscopic. the sample in the reservoir is transferred to freshly cleaved mica surface by a pipette. After 15 min, the sample is washed with Milli-Q water three times and dried in air. The control experiment is conducted by injecting only buffer into the channel. The mixed buffer solution after the treatment of the same electric field is used as the blank. The AFM images were taken in air in tapping mode on Nanoscope III(a) (Veeco. Instrument Inc.) equipped with EV scanner. RFESP tip (Veeco. Instrument Inc.) is used in the experiment. The resonant frequency of the cantilever is about 90 kHz.



Figure S1: (A) applied voltage for sample injection (unchanged); (B) applied voltage for sample to form complex at 125 V/cm. "B" in the figure represents "buffer". The running electric field can be tuned by multiplying all the voltages by twice (250 V/cm), three times (375V/cm), or four times (495V/cm).



Figure S2: Complex of DNA and PLL prepared by mixing in vials (A) or in microchannel (B) without electric field. In panel A, the +/- charge ratios or samples 2 to 6 are 0.6, 1.2, 1.8, 2.5, and 3.1.



Figure S3: AFM images of the complex migrate out of the channel.



Figure S4: Time dependence of the front particle mobility at 125 V/cm.



Figure S5. migration of free DNA under 125 V/cm.



Figure S6: characteristic time  $\tau$  decreases with electric field strength.

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- 2. E. Wernersson, J. Heyda, A. Kubíčková, T. s. Křížek, P. Coufal and P. Jungwirth, *The Journal of Physical Chemistry B*, 2010, **114**, 11934-11941.